62

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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

**(51) International Patent Classification 6 :**
**C08L 23/10, C08F 10/00**

**A1**

**(11) International Publication Number:** **WO 98/45368**

**(43) International Publication Date:** 15 October 1998 (15.10.98)

**(21) International Application Number:** PCT/JP98/01603

**(22) International Filing Date:** 7 April 1998 (07.04.98)

**(30) Priority Data:**
9/92387 10 April 1997 (10.04.97) JP

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**(81) Designated States:** KR, US, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).

**Published**
*With international search report.*

**(54) Title:** OLEFIN (CO–)POLYMER COMPOSITIONS AND METHOD OF PRODUCING THE SAME

**(57) Abstract**

The invention provides an olefin (co–)polymer composition having a high melt tension, high crystallization temperature and high rigidity. A method of producing the composition comprises: preparing a preactivated catalyst by forming a small amount of high molecular weight polyolefin in advance; further adding a specific catalyst component to the preactivated catalyst; and then performing main polymerization using the combined catalyst. The olefin (co–)polymer composition mainly comprises 100 weight parts of an olefin (co–)polymer having an intrinsic viscosity [$\eta_b$] of 0.2 to 10 dl/g measured in tetralin at 135 °C, and it is produced by (co–)polymerizing olefins using a polymerization catalyst containing: (a) 0.01 to 5 weight parts of an olefin (co–)polymer having an intrinsic viscosity [$\eta_a$] of 15 to 100 dl/g measured in tetralin at 135 °C; and (b) catalyst components (1), (2 and 3) as follows: the component (1) of a titanium trichloride composition (III), the catalyst component (2) of an organic aluminum compound ($A_2$), and the catalyst component (3) of at least one compound selected from the group consisting of aromatic carboxylic acid esters (E) and organic silicon compounds (S) containing either a Si–O–C group or a mercapto group.

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# DESCRIPTION

# OLEFIN (CO-)POLYMER COMPOSITIONS AND METHOD OF PRODUCING THE SAME

## TECHNICAL FIELD

The present invention relates to olefin (co-)polymers, particularly to an olefin (co-)polymer composition having high rigidity, high melt tension and high crystallization temperature, and also excellent thermal stability, and to a method of producing the same.

## BACKGROUND OF THE INVENTION

Olefin (co-)polymers such as polypropylene and polyethylene are excellent in mechanical properties, chemical resistance and the like, and are also very useful in view of balance with economy, so that they have been widely used in various molding fields. However, because such olefin (co-)polymers have a low melt tension and a low crystallization temperature, they exhibit poor moldability in molding methods such as blow molding, foaming, extrusion molding or the like. Also, in various other molding methods, molding productivity at high speed is also limited.

In order to improve the melt tension and the crystallization temperature of an olefin (co-)polymer, methods of reacting an organic peroxide and a crosslinking assistant with polypropylene in the molten state (e.g. Japanese Published Unexamined Patent Applications No. SHO 59-93711 and No. SHO 61-152754), and methods of producing polypropylene

having free-end long branches but containing no gel by reacting semi-crystalline polypropylene with a peroxide having a low decomposition temperature (e.g. Japanese Published Unexamined Patent Application No. HEI 2-298536) have been proposed.

Other methods also have been proposed to improve the melt viscoelasticity such as melt tension. For example, compositions comprising a mixture of polyethylene or polypropylene having different intrinsic viscosities or molecular weights, and a method of producing such compositions by multi-step polymerization, have been proposed.

For example, the following methods have been proposed: a method of mixing 2 to 30 weight parts of ultra high molecular weight polypropylene with 100 weight parts of normal polypropylene, and extruding the mixture at a temperature from the melting point to 210 ℃ (Japanese Published Examined Patent Application No. SHO 61-28694), an extruded sheet comprising two different polypropylene compositions having different molecular weights with a ratio of the limiting viscosity of not less than 2, which is produced by multi-step polymerization (Japanese Published Examined Patent Application No. HEI 1-12770), a method of producing a polyethylene composition comprising three different polyethylene compositions having different viscosity average molecular weights by melt mixing or multi-step polymerization (Japanese Published Examined Patent Application No. SHO 62-61057), a method of polymerizing an ultra high molecular weight polyethylene having a limiting viscosity of not less than 20 dl/g in an amount of less than 0.05 to 1 weight % by a multi-step

polymerization using a highly actiated titanium-vanadium solid catalyst composition (Japanese Published Examined Patent Application No. HEI 5-79683), and a method of polymerizing an ultra high molecular weight polyethylene having a limiting viscosity of not less than 15 dl/g in an amount of 0.1 to 5 weight % by multi-step polymerization using a polymerization reactor with a special arrangement using a highly activated titanium catalyst composition preliminarily treated by polymerization of 1-butene or 4-methyl-1-pentene (Japanese Published Examined Patent Application No. HEI 7-8890).

Furthermore, a method of producing polypropylene having high melt tension by polymerizing propylene using a preactiaved catalyst prepared by preliminarily polymerizing ethylene and a polyene compound using a supported Ti containing solid catalyst component and an organic aluminum compound catalyst component (Japanese Published Unexamined Patent Application No. HEI 5-222122), and a method of producing a linear low density polyethylene (LLDPE) having high melt tension by using a preactivated catalyst containing polyethylene having a limiting viscosity of not less than 20 dl/g prepared by preliminarily polymerizing only ethylene using the same catalyst components as in the former method as described above (Japanese Published Unexamined Patent Application No. HEI 4-55410), have been proposed.

## SUMMARY OF THE INVENTION

In the various compositions and methods of producing the same proposed as mentioned above, the olefin (co-)polymers as obtained had

improved melt tension to some degree. However, further improvement is still needed with respect to problems such as residual odor due to the use of crosslinking assistants, crystallization temperature, thermal stability and the like

Furthermore, in the multi-step polymerization method wherein a process of producing a high molecular weight polyolefin is incorporated in a normal process of (co-)polymerizing olefins in main polymerization, it is difficult to control (co-)polymerization of olefin so as to form a trace amount of the high molecular weight polyolefin, and furthermore, a low polymerization temperature also is required to produce a polyolefin having a sufficiently large molecular weight. Therefore, conversion of the process is required, and furthermore, the productivity of the final polyolefin composition is reduced.

In the method of preliminary polymerizing a polyene compound, it is necessary to prepare it separately. Furthermore, in the method of preliminarily polymerizing polyethylene, because the preliminarily polymerized polyethylene is not uniformly dispersed in the final product of polyolefin composition, further improvement is still required with respect to the stability of the polyolefin composition.

As described above, such olefin (co-)polymers produced according to the conventional techniques are not sufficiently improved in melt tension and crystallization temperature, and moreover, there are problems to be improved with respect to residual odor and thermal stability. Furthermore, in producing such olefin (co-)polymers, it is required to improve the

productivity.

It is an object of the present invention to provide an olefin (co-)polymer composition having high melt tension and high crystallization temperature, which is suitable for blow molding, foaming and extrusion molding, and also allows high speed forming in other various molding methods, and to provide a method of producing the same.

As a result of ardent research to achieve the above objects, the inventors discovered that an olefin (co-)polymer composition having high rigidity, as well as high melt tension and high crystallization temperature, can be obtained by performing main (co-)polymerization of olefins using a catalyst prepared by a preliminary activation of a particular catalyst for polyolefin production by (co-)polymerizing olefins to form a trace amount of an olefin (co-)polymer having a specific intrinsic viscosity before main (co-)polymerization, and further adding a particular catalyst component thereto.

In order to attain the above-mentioned object, the present invention provides an olefin (co-)polymer composition mainly comprising:

(a) 0.01 to 5 weight parts of an olefin (co-)polymer (a) having an intrinsic viscosity $[\eta_a]$ of 15 to 100 dl/g measured in tetralin at 135℃; and

(b) 100 weight parts of an olefin (co-)polymer (b) having an intrinsic viscosity $[\eta_b]$ of 0.2 to 10 dl/g measured in tetralin at 135℃, which is produced by (co-)polymerizing olefins using a polymerization catalyst comprising catalyst components (1), (2) and (3) as follows:

Catalyst Component (1): a titanium trichloride composition (Ⅲ)

Catalyst Component (2): an organic aluminum compound ($A_2$)

Catalyst Component (3): at least one compound selected from the group consisting of aromatic carboxylic acid esters (E), and organic silicon compounds (S) that contain at least one organic group selected from the group consisting of a Si-O-C group and a mercapto group.

The expression "mainly comprising" as used herein means that the olefin (co-)polymer composition of the present invention contains a composition comprising (a) and (b) at not less than 30 %, preferably not less than 40 %, more preferably not less than 50 %.

In the above-mentioned polymer composition of the present invention, the titanium trichloride composition (Ⅲ) is preferably a composition produced by reacting titanium tetrachloride with either an organic aluminum compound ($A_1$) or a reaction product (Ⅰ) of the organic aluminum compound ($A_1$) and an electron donor ($B_1$) to form a solid product (Ⅱ), and then reacting the (Ⅱ) with an electron donor ($B_2$) and an electron acceptor.

Furthermore, the olefin (co-)polymer composition according to the present invention preferably has a relationship between the melt tension (MS) at 230 ℃ and the melt flow rate (MFR) measured under a load of 21.18 N at 230 ℃ as follows:

$$\log(\mathrm{MS}) > -0.76 \times \log(\mathrm{MFR}) + 0.45$$

By satisfying the above relationship, the composition of the present invention is provided with high melt tension and good moldability. (For

example, in large size moldings, molding of a resin composition cannot be performed if the melt tension is too low. Furthermore, a resin composition needs a high MFR value to exhibit high speed moldability.) In general, as the melt tension of a resin composition is high, it has a low MFR value and therefore exhibit insufficient moldability.

Although there is no specific upper limit, if the melt tension is too high, the moldability of the composition is deteriorated. Therefore, it is preferably within a range of:

$$-0.76 \times \log(\mathrm{MFR}) + 2.00 > \log(\mathrm{MS}) > -0.76 \times \log(\mathrm{MFR}) + 0.45.$$

More preferably, it is within a range of:

$$-0.76 \times \log(\mathrm{MFR}) + 1.80 > \log(\mathrm{MS}) > -0.76 \times \log(\mathrm{MFR}) + 0.54.$$

In the polymer composition of the present invention, the olefin (co-)polymer (a) is preferably either an ethylene homopolymer or an ethylene-olefin (co-)polymer containing not less than 50 weight % of ethylene polymerization units.

In the polymer composition of the present invention, the olefin (co-)polymer (b) is preferably either a propylene homopolymer or a propylene-olefin copolymer containing not less than 50 weight % of propylene polymerization units.

In the polymer composition of the present invention, the olefin (co-)polymer composition preferably has a ratio (Mw/Mn) of the weight average molecular weight (Mw) to the numerical average molecular weight (Mn) in the range of 6 to 30.

Furthermore, a method of producing an olefin (co-)polymer composition

of the present invention comprises: performing a preliminary activation of a polymerization catalyst comprising catalyst components (1) and (2) below by (co-)polymerizing olefins to form 0.01 to 5,000 g of an olefin (co-)polymer (a) having an intrinsic viscosity [$\eta_a$] of 15 to 100 dl/g measured in tetralin at 135℃ per 1 g of the catalyst component (1) to prepare a preactivated catalyst; combining the preactivated catalyst further with a catalyst component (3) below to prepare a polymerization catalyst; and then performing main (co-)polymerization of olefins using the polymerization catalyst to form an olefin (co-)polymer (b) having an intrinsic viscosity [$\eta_b$] of 0.2 to 10 dl/g measured in tetralin at 135℃. This produces an olefin (co-)polymer composition mainly comprising:

(a) 0.01 to 5 weight parts of an olefin (co-)polymer (a) having an intrinsic viscosity [$\eta_a$] of 15 to 100 dl/g measured in tetralin at 135℃; and

(b) 100 weight parts of an olefin (co-)polymer (b) having an intrinsic viscosity [$\eta_b$] of 0.2 to 10 dl/g measured in tetralin at 135℃.

Catalyst Component (1): a titanium trichloride composition (III)

Catalyst Component (2): an organic aluminum compound ($A_2$)

Catalyst Component (3): at least one compound selected from the group consisting of aromatic carboxylic acid esters (E), and organic silicon compounds (S) that contain at least one organic group selected from the group consisting of a Si-O-C group and a mercapto group.

In the method of the present invention, the titanium trichloride composition (III) is preferably a composition produced by reacting titanium

tetrachloride with either an organic aluminum compound ($A_1$) or a reaction product ( I ) of the organic aluminum compound ($A_1$) and an electron donor ($B_1$) to form a solid product ( II ), and then reacting the ( II ) with an electron donor ($B_2$) and an electron acceptor.

Furthermore, in the method of the present invention, (co-)polymerization of olefins also can be performed using a preactivated catalyst further combined with an additional catalyst component (2) below.

Catalyst Component (2): an organic aluminum compound ($A_2$)

Furthermore, in the method of the present invention, the olefin (co-)polymer composition obtained preferably has a relationship between the melt tension (MS) at 230 ℃ and the melt flow rate (MFR) measured under a load of 21.18 N at 230 ℃ as follows:

$\log(MS) > -0.76 \times \log(MFR) + 0.45$

In the method of the present invention, the olefin (co-)polymer (a) is preferably either an ethylene homopolymer or an ethylene-olefin (co-)polymer containing not less than 50 weight % of ethylene polymerization units.

In the method of the present invention, the olefin (co-)polymer (b) is preferably either a propylene homopolymer or a propylene-olefin copolymer containing not less than 50 weight % of propylene polymerization units.

In the method of the present invention, it is possible to perform an additional preliminary activation by (co-)polymerizing olefins before or before and after the preliminary activation to form 0.01 to 100 g of an olefin (co-)polymer (aa) having an intrinsic viscosity of $[\eta_{aa}]$ lower than the

intrinsic viscosity [ $\eta_a$] of the olefin (co-)polymer (a) formed by the preliminary activation per 1 g of the catalyst component (1).

In the method of the present invention, the olefin (co-)polymer (aa) is preferably either a propylene homopolymer or a propylene-olefin copolymer containing not less than 50 weight % of propylene polymerization units.

In the method of the present invention, the olefin (co-)polymer composition (b) preferably has a ratio (Mw/Mn) of the weight average molecular weight (Mw) to the numerical average molecular weight (Mn) in the range of 6 to 30.

## BRIEF DESCRIPTION OF THE DRAWING

Figure 1 is a flow sheet explaining a method of producing an olefin (co-)polymer composition of the present invention.

## DESCRIPTION OF THE PREFERRED EMBODIMENTS

The term "preliminary activation" used in the present specification refers to (co-)polymerization of olefins in a trace amount (normally not more than 5 weight % of the quantity formed in main (co-)polymerization), which is carried out before main (co-)polymerization of olefins. The preliminary activation uses a catalyst for olefin (co-)polymerization prepared by combining a titanium trichloride composition as the transition metal compound catalyst component (1) for olefin (co-)polymerization with the catalyst component (2) as an activator to promote the olefin polymerization performance by acting on the titanium trichloride composition. After the preliminary activation is carried out, the surface of the catalyst component (1) is covered with an olefin (co-)polymer.

The olefin (co-)polymer (a) constituting the component (a) of the olefin (co-)polymer composition of the present invention has an intrinsic viscosity [$\eta_a$] of 15 to 100 dl/g measured in tetralin at 135 ℃, and it may be either a homopolymer or a copolymer of olefins having 2 to 12 carbon atoms. However, it is preferably either an ethylene homopolymer or an ethylene-olefin copolymer containing not less than 50 weight % of ethylene polymerization units, more preferably either an ethylene homopolymer or an ethylene-olefin copolymer containing not less than 70 weight % of ethylene polymerization units, and particularly more preferably either an ethylene homopolymer or an ethylene-olefin copolymer containing not less than 90 weight % of ethylene polymerization units. These (co-)polymers may be used either alone or in combinations of two or more types.

In order to improve the melt tension and the crystallization temperature of the olefin (co-)polymer composition finally obtained, to improve dispersion of the olefin (co-)polymer (a) in the olefin (co-)polymer (b) formed by main polymerization, and further to improve the efficiency of the production, the olefin (co-)polymer (a) suitably has an intrinsic viscosity [$\eta_a$] of 15 to 100 dl/g, preferably 17 to 50 dl/g.

Although the types of the olefins constituting the olefin (co-)polymer component (a) are not particularly limited, olefins having 2 to 12 carbon atoms are preferably used. Examples include ethylene, propylene, 1-butene, 1-pentene, 1-hexene, 1-octene, 1-decene, 4-methyl-1-pentene, 3-methyl-1-pentene or the like, and particularly ethylene is suitably used as a main monomer. These olefins may be used either alone or in combinations of two

or more types.

Although the density of the olefin (co-)polymer (a) is not particularly limited, it is preferably in the range of about 880 to 980 g/l.

The olefin (co-)polymer (b) constituting the component (b) of the olefin (co-)polymer composition of the present invention has an intrinsic viscosity $[\eta_b]$ of 0.2 to 10 dl/g measured in tetralin at 135 ℃, and it can be either a homopolymer or a copolymer of olefins having 2 to 12 carbon atoms. It is preferably either a propylene homopolymer, or a propylene-olefin random copolymer or a propylene-olefin block copolymer containing not less than 50 weight % of propylene polymerization units, and more preferably either a propylene homopolymer, a propylene-olefin random copolymer containing not less than 90 weight % of propylene polymerization units, or a propylene-olefin block copolymer containing not less than 70 weight % of propylene polymerization units. These (co-)polymers may be used either alone or in combination of two or more types.

In order to provide good mechanical properties and good moldability of the olefin (co-)polymer composition finally obtained, the olefin (co-)polymer (b) used has an intrinsic viscosity $[\eta_b]$ of 0.2 to 10 dl/g, preferably 0.5 to 8 dl/g.

Although the types of the olefins constituting the olefin (co-)polymer component (b) are not particularly limited, olefins having 2 to 12 carbon atoms are preferably used. Examples include ethylene, propylene, 1-butene, 1-pentene, 1-hexene, 1-octene, 1-decene, 4-methyl-1-pentene, 3-methyl-1-pentene or the like, and particularly propylene is suitably used as a main

monomer. These olefins may be used either alone or in combinations of two or more types.

With regard to the molecular weight distribution of the olefin (co-)polymer (b), in order to finally obtain an olefin (co-)polymer composition having improved rigidity, the ratio (Mw/Mn) of the weight average molecular weight (Mw) to the numerical average molecular weight (Mn) is preferably in the range of 6 to 30, and more preferably in the range of 7 to 30.

The ratio (Mw/Mn) of the weight average molecular weight (Mw) to the numerical average molecular weight (Mn) is a measure of the molecular weight distribution. As the ratio (Mw/Mn) becomes larger, the molecular weight distribution becomes wider, and as the ratio (Mw/Mn) becomes smaller, the molecular weight distribution becomes narrower.

The above-mentioned ratio (Mw/Mn) of the weight average molecular weight (Mw) to the numerical average molecular weight (Mn) is calculated based on the results of the weight average molecular weight (Mw) and the numerical average molecular weight (Mn), measured by means of gel permeation chromatography (GPC) according to a method as follows: using an o-dichlorobenzene solution containing 0.05 weight % of polymer, and using a mixed polystyrene gel column (e.g. PSKgel GMH6-HT produced by TOSOH CORPORATION), measurement is conducted at a temperature of 135℃. For example, GPC-150C produced by Waters is used as the measuring apparatus.

It is preferable that the olefin (co-)polymer (b) has a relatively high stereoregularity so that the rigidity of the olefin (co-)polymer composition

finally obtained can be improved.

For example, when polypropylene is used as the olefin (co-)polymer (b), the polypropylene used suitably has an isotactic pentad tacticity (mmmm) of 0.93 to 0.99, preferably 0.94 to 0.99, particularly more preferably 0.95 to 0.99 measured by means of $^{13}$C-NMR (nuclear magnetic resonance spectrum).

The term "isotactic pentad tacticity" (mmmm) used herein refers to isotactic tacticity in a polypropylene molecular chain in a pentad unit, measured by means of $^{13}$C-NMR, which was proposed by A. Zambelli et al. (Macromolecules 6,925 (1973)). The assignment of the peak in the measuring spectrum is determined according to a method proposed by A. Zambelli et al (Macromolecules 8,687 (1975)). In this method, the assignment is determined by a measurement using a mixture solution comprising 20 weight % of polymer, and o-dichlorobenzene and bromobenzene at a ratio of 8:2 by weight, under conditions of at 67.20MHz and at a temperature of 130℃. For example, JEOL-GX270NMR produced by JEOL, Ltd. is used as the measuring apparatus.

The olefin (co-)polymer (b) as the component (b) of the present invention is obtained by (co-)polymerizing olefins using a catalyst comprising the catalyst components (1), (2) and (3) as follows:

Catalyst Component (1): a titanium trichloride composition (Ⅲ)

Catalyst Component (2): an organic aluminum compound ($A_2$)

Catalyst Component (3): at least one compound selected from the group consisting of aromatic carboxylic acid esters (E), and organic silicon compounds (S) that contain at least one organic group selected from the

group consisting of a Si-O-C group and a mercapto group.

The titanium trichloride composition (Ⅲ) as the catalyst component (1) can be a known titanium trichloride composition as the transition metal compound catalyst component contained in a so-called Ziegler-Natta catalyst used for olefin polymerization. However, such a titanium trichloride composition produced according to a process as described below is preferably used so that the composition of the present invention obtained has improved rigidity.

Such a titanium trichloride composition (Ⅲ) as the catalyst component (1) is prepared as follows: First, a solid product (Ⅱ) is produced either by (i) reacting an organic aluminum compound ($A_1$) with titanium tetrachloride or by (ii) reacting the reaction product (Ⅰ), which is formed by reacting ($A_1$) and an electron donor ($B_1$), with titanium tetrachloride. According to the process (ii), a more desirable catalyst component (1) can be finally obtained. The process (ii) is disclosed in the specification of Japanese Published Unexamined Patent Application No. SHO 56-110707 as follows: That is, the reaction between an organic aluminum compound ($A_1$) and an electron donor ($B_1$) is performed in a solvent ($D_1$) at a temperature of −20 ℃ to 200 ℃, preferably −10 to 100 ℃, for a period of from 30 seconds to 5 hours. There is no limitation in the order of adding ($A_1$), ($B_1$) and ($D_1$), and furthermore, the ratio of the quantities for each component used is suitably in the range of 0.1 to 8 mole, preferably 1 to 4 mole of the electron donor ($B_1$), and 0.5 to 5 liters, preferably 0.5 to 2 liters of the solvent ($D_1$) respectively per one mole of the organic aluminum compound ($A_1$). An aliphatic hydrocarbon is

preferably used as the solvent. Thus the reaction product ( I ) is obtained.

The reaction between the reaction product ( I ) and titanium tetrachloride is carried out at a temperature of 0 to 200 ℃, preferably 10 to 90 ℃, for a period of from 5 minutes to 8 hours. Although it is preferable not to use any solvent, an aliphatic or aromatic hydrocarbon may be used as a solvent. Although the ( I ), titanium tetrachloride and a solvent may be mixed in any order, it is preferable to complete the mixing of the total amount within 5 hours, and after completing the mixing of the total amount, further reaction is preferably continued at a temperature of 10 to 90 ℃ for up to 8 hours. The amount of each component used in the reaction is as follows: 0 to 3,000 ml of the solvent per one mole of titanium tetrachloride, and with regard to the reaction product ( I ), the ratio (Al/Ti) of the number of Al atoms contained in ( I ) to the number of Ti atoms contained in titanium tetrachloride being 0.05 to 10, preferably 0.06 to 0.2. After completing the reaction, the liquid phase portion is separated and removed from the mixture by filtration or decantation, followed by repeated washing with a solvent. The solid product (II) thus obtained may be used in the next step as a suspension in the solvent, or taken out after being dried and used as a solid substance.

Then, the solid product (II) is allowed to react with an electron donor ($B_2$) and an electron acceptor. Although this reaction may be carried out without using any solvent, more desirable results can be obtained when an aliphatic hydrocarbon is used as a solvent. The quantities of each used are 10 to 1,000 g, preferably 50 to 200 g of ($B_2$), 10 to 1,000 g, preferably 20 to 500

g of the electron donor, and 0 to 3,000 ml, preferably 100 to 1,000 ml of the solvent respectively per 100 g of the solid product (II). These three or four substances are mixed at a temperature of −10 to 40 ℃ for a period of from 30 seconds to 60 minutes, and it is desired that the mixture is allowed to react at a temperature of 40 to 200℃, preferably at 50 to 100℃ for a period of from 30 minutes to 5 hours. The order of mixing the solid product (II), ($B_2$), an electron acceptor and a solvent is not limited. The ($B_2$) and an electron acceptor may be allowed to react with each other previously before being mixed with the solid product (II). In this case, the ($B_2$) and the electron acceptor are allowed to react with each other at a temperature of 10 to 100 ℃ for a period of from 30 minutes to 2 hours, and then cooled down to a temperature at 40 ℃ or lower. After the reaction of the solid product (II), the ($B_2$) and the electron acceptor is completed, the liquid phase of the reacted mixture is separated and removed by filtration or decantation, then washing with a solvent is repeated, and then unreacted liquid materials are removed, thus obtaining a titanium trichloride composition (III). The titanium trichloride composition (III) may be provided for the next use either as a solid substance taken out after being dried, or as a suspension in the solvent.

The organic aluminum compound ($A_1$) used in the production of titanium trichloride compound (III) of the present invention is an organic aluminum compound represented by the following formula:

$$AlR^1_pR^2_qX_{3-(p+q)}$$

wherein $R^1$ and $R^2$ respectively represent hydrocarbon groups such as an

alkyl group, cycloalkyl group, aryl group or the like, or an alkoxyl group, X represents a halogen atom, and p and q are respectively arbitrary integers satisfying a formula $0 < p+q \leqq 3$.

Examples of the organic aluminum compound ($A_1$) include trialkyl aluminums such as trimethyl aluminum, triethyl aluminum, tri-n-propyl aluminum, tri-n-butyl aluminum, tri-i-butyl aluminum, tri-n-hexyl aluminum, tri-i-hexyl aluminum, tri-n-octyl aluminum and the like, dialkyl aluminum monohalides such as diethyl aluminum chloride, di-n-propyl aluminum chloride, di-i-butyl aluminum chloride, diethyl aluminum bromide, diethyl aluminum iodide and the like, dialkyl aluminum hydrides such as diethyl aluminum hydride and the like, alkyl aluminum sesquihalide such as ethyl aluminum sesquichloride and the like, monoalkyl aluminum dihalides such as ethyl aluminum dichloride and the like. Furthermore, alkoxyalkyl aluminums such as diethoxy monoethyl aluminum or the like also can be used as the organic aluminum compound ($A_1$). Among these examples, trialkyl aluminums or dialkyl aluminum monohalides are preferably used. Furthermore, these organic aluminum compounds also can be used in combinations of two or more types, including different types if desired.

Ethers are mainly used as the electron donor ($B_1$) in the present invention, but other electron donors also can be used additionally as long as the effect of the present invention is not impeded. Examples of ethers include diethyl ether, di-n-propyl ether, di-n-butyl ether, di-i-amyl ether, di-n-pentyl ether, di-n-hexyl ether, di-i-hexyl ether, di-n-octyl ether, di-n-dodecyl ether, diphenyl ether, ethylene glycol monoethyl ether,

tetrahydrofuran and the like. Furthermore, examples of electron donors that can be used together with ethers include organic compounds having any of oxygen, nitrogen, sulfur and phosphorus atoms in the molecule, such as alcohols, phenols, esters, aldehydes, fatty acids, aromatic acids, ketones, nitriles, amines, amides, ureas, isocyanates, azo-compounds, phosphines, phosphites, phosphinates, thioethers, thioalcohols, organic silicon compounds having a Si-O-C group or a mercapto group, and the like. These electron donors also can be used as a mixture thereof.

Halides of elements from the Group III (e.g. La), Group IV (e.g. Ti, Zr), Group V (e.g. V), Group VI (e.g. Cr), Group XIII (e.g. Al), Group XIV (e.g. Si, Sn), and Group XV (e.g. P, Sb) in the periodic table are representative of the electron donor used in the present invention. Examples include anhydrous aluminum chloride, silicon tetrachloride, stannous chloride, stannic chloride, titanium tetrachloride, zirconium tetrachloride, phosphorus trichloride, phosphorus pentachloride, vanadium tetrachloride, antimony pentachloride and the like. These examples also can be used as a mixture thereof. Among these, titanium tetrachloride is most preferably used.

Examples of the solvent ($D_1$) include aliphatic hydrocarbons such as n-pentane, n-hexane, n-heptane, n-octane, i-octane and the like, alicyclic hydrocarbons such as cyclohexane, metylcyclohexane and the like, hydrocarbon halides such as carbon tetrachloride, chloroform, dichloroethane, trichloroethylene, tetrachloroethylene and the like, aromatic hydrocarbons such as benzene, toluene, xylene, ethylbenzene and the like,

and aromatic hydrocarbon halides such as monochlorobenzene, chloroethylbenzene, dichlorobenzene and the like.

The organic aluminum compound ($A_2$) as the catalyst component (2) of the present invention can be the same compound as the organic aluminum compound ($A_1$) used in producing the titanium trichloride composition (III). Moreover, the organic aluminum compound ($A_2$) can be either the same or a different compound as the organic aluminum compound ($A_1$) used in producing the titanium trichloride composition (III).

As the catalyst component (3) of the present invention, one or more compounds selected from the group consisting of aromatic carboxylic acid esters (E) and organic silicon compounds (S) having a Si-O-C group or a mercapto group are used.

Examples of aromatic carboxylic acid esters (E) include ethyl benzoate, propyl benzoate, butyl benzoate, octyl benzoate, 2-ethylhexyl benzoate, toluic acid methyl ester, toluic acid ethyl ester, toluic acid 2-ethylhexyl ester, anisic acid methyl ester, anisic acid ethyl ester, anisic acid propyl ester, ethyl cinnamate, naphthoic acid butyl ester, naphthoic acid propyl ester, naphthoic acid butyl ester, naphthoic acid 2-ethylhexyl ester, ethyl phenylacetate and the like.

Examples of organic silicon compounds (S) having a Si-O-C group or a mercapt group include organic silicon compounds having a Si-O-C group such as methyltrimethoxysilane, vinyltrimethoxysilane, allyltrimethoxysilane, phenyltrimethoxysilane, dimethyldimethoxysilane, methylphenyldimethoxysilane, diphenyldimethoxysilane,

trimethylmethoxysilane, triphenylmethoxysilane, tetraethoxysilane, methyltriethoxysilane, ethyltriethoxysilane, vinyltriethoxysilane, propyltriethoxysilane, allyltriethoxysilane, pentyltriethoxysilane, phenyltriethoxysilane, n-octyltriethoxysilane, n-octadecyltriethoxysilane, 6-triethoxysilane-2-norbornene, dimethyldiethoxysilane, diethyldiethoxysilane, diphenyldiethoxysilane, trimethylethoxysilane, triethylethoxysilane, triphenylethoxysilane, allyloxytrimethylsilane, methyl-tri-i-propoxysilane, dimethyl-di-i-propoxysilane, trimethyl-i-propoxysilane, tetra-n-butoxysilane, methyl-tri-n-butoxysilane, tetra(2-ethylbutoxy)silane, methyltriphenoxysilane, dimethyldiphenoxysilane, trimethylphenoxysilane, trimethoxysilane, triethoxysilane, triethoxychlorosilane, tri-i-propoxychlorosilane, tri-n-butoxychlorosilane, tetraacetoxysilane, methyltriacetoxysilane, ethyltriacetoxysilane, vinyltriacetoxysilane, methyldiacetoxysilane, diacetoxymethylvinylsilane, dimethyldiacetoxysilane, methylphenyldiacetoxysilane, diphenyldiacetoxysilane, trimethylacetoxysilane, triethylacetoxysilane, phenyldimethylacetoxysilane, triphenylacetoxysilane, bis-(trimethylsilyl)adipate, trimethylsilylbenzoate, triethylsilylbenzoate and the like; and

further include organic silicon compounds having a mercapto group such as mercaptomethyltrimethylsilane,

2-mercaptoethyltrimethylsilane,

3-mercaptopropyltrimethylsilane, 4-mercapto-n-butyltrimethylsilane, mercaptomethyltriethylsilane, 2-mercaptoethyltriethylsilane,

3-mercaptopropyltriethylsilane, 1-mercaptoethyltrimethylsilane,

3-mercaptopropyldimethylphenylsilane, 3-mercaptopropylethylmethylphenylsilane, 4-mercaptobutyldiethylphenylsilane, 3-mercaptopropylmethyldiphenylsilane and the like; and

still further include organic silicon compounds having a Si-O-C group and a mercapto group such as mercaptomethyltrimethoxysilane, mercaptomethyldimethoxymethylsilane, mercaptomethylmethoxydimethylsilane, mercaptomethyltriethoxysilane, mercaptomethyldiethoxymethylsilane, mercaptomethylethoxydimethylsilane, 2-mercaptoethyltrimethoxysilane, 3-mercaptopropyltrimethoxysilane, dimethoxy-3-mercaptopropylmethylsilane, 3-mercaptopropyltriethoxysilane, diethoxy-3-mercaptopropylmethylsilane, mercaptomethyldimethyl-2-phenylethoxysilane, 2-mercaptoethoxytrimethylsilane, 3-mercaptopropoxytrimethylsilane and the like; and

still further include organic silicon compounds having a Si-O-C group and an amino group such as 3-aminopropyltrimethoxysilane, 3-aminopropyltriethoxysilane, 3-aminopropyldiethoxymethylsilane, 3-aminopropyldimethylethoxysilane, 3-aminophenoxydimethylvinylsilane, 4-aminophenoxydimethylvinylsilane, 2-aminoethylaminomethyltrimethoxysilane, 3-(2-aminoethylaminopropyl)dimethoxymethylsilane, 2-aminoethylaminomethylbenzyloxydimethylsilane, 2-aminoethylpropyl-tri-methoxysilane, and the like.

The olefin (co-)polymer (b) used in the present invention can be obtained by (co-)polymerizing olefins using a catalyst containing the above-mentioned catalyst components (1) to (3).

At this time, the quantities of the catalyst components used are in a ratio of the organic aluminum compound ($A_2$) as the catalyst component (2) to the Ti atoms contained in the titanium trichloride composition (III) as the catalyst component (1) that is 0.1 to 500, preferably 0.1 to 200 by mole, and furthermore, in a ratio of the aromatic carboxylic acid esters (E) or organic silicon compounds (S) having a Si-O-C group or a mercapto group as the catalyst component (3) to the Ti atoms contained in the titanium trichloride composition (III) as the catalyst component (1) that is 0.2 to 50, preferably 0.5 to 20 by mole.

It is desirable that the amount of the catalyst component (3) used is not less than the above-mentioned lower limit so as to improve crystallinity and to provide larger distribution of the molecular weight, and also preferably not exceeding the above-mentioned upper limit so as to improve polymerization activity.

The olefin (co-)polymer (b) used in the composition of the present invention can be obtained by (co-)polymerizing olefins using a particular catalyst containing the respective catalyst components thus combined. The (co-)polymerization of olefins can be performed by a known (co-)polymerization process for olefins. Examples of such a (co-)polymerization process include slurry polymerization, in which olefin is polymerized in an inert solvent, such as aliphatic hydrocarbons like butane,

pentane, hexane, heptane or isooctane, alicyclic hydrocarbons like cyclopentane, cyclohexane or methyl cyclohexane, aromatic hydrocarbons like toluene, xylene or ethylbenzene, or gasoline fraction or hydrogenized diesel oil fraction; bulk polymerization, in which the olefin itself is used as the solvent; gas phase polymerization, in which olefins are (co-)polymerized in the gas phase; and solution polymerization, in which the polyolefin formed by the (co-)polymerization is in liquid state. The combinations of two or more types of these processes also can be used.

In polymerizing olefins, the components of the above-mentioned specific catalyst may be mixed in an inert solvent in advance and then provided to the polymerization reaction system, or each component may be separately provided to the polymerization reaction system. Moreover, it is also preferable to previously perform preliminary activation, in which a small amount of olefin, specifically about 0.01 to 500 g of olefin with respect to 1 g of the catalyst component (1), is polymerized in an inert solvent in the presence of at least the catalyst component (1) and the catalyst component (2) to produce a preactivated catalyst, before main polymerization of olefin is performed. The olefin (co-)polymer (b) thus obtained is of fine particulate form.

In the preliminary activation as mentioned above, $\alpha$-olefin having 2 to 12 carbon atoms can be preferably used, and examples include ethylene, propylene, butene, pentene, hexene, octene, 4-methyl-1-pentene and the like. Particularly, ethylene, propylene or 4-methyl-1-pentene is preferably used.

Thus, a particular catalyst or a preliminarily activated particular

catalyst, which is prepared for use in the present invention, is used in (co-)polymerizing olefins by the above-mentioned polymerization method. However, the same polymerization conditions as in (co-)polymerization of olefins using a known Ziegler type catalyst are usually applied in the propylene polymerization. Therefore, the polymerization is carried out at a temperature of 0 to 150 ℃, preferably 20 to 120 ℃, more preferably 40 to 100 ℃, under a pressure ranging from the atmospheric pressure to 7 MPa, preferably from 0.2 MPa to 5 MPa, in the presence of hydrogen as a molecular weight controlling agent, usually for about one minute to 20 hours, to form an olefin (co-)polymer (b) having an intrinsic viscosity $[\eta_b]$ of 0.2 to 10 dl/g measured in tetralin at 135 ℃.

After the (co-)polymerization of olefin is completed, after-treatment is carried out with known processes such as catalyst deactivation, removing catalyst residue, drying and so forth as needed, and the olefin (co-)polymer (b) having an intrinsic viscosity $[\eta_b]$ of 0.2 to 10 dl/g measured in tetralin at 135 ℃ used in the present invention is obtained.

Furthermore, it is preferable that the olefin (co-)polymer (b) has a ratio (Mw/Mn) of the weight average molecular weight (Mw) to the numerical average molecular weight (Mn) in the range of 6 to 30, more preferably 7 to 30 so as to improve the rigidity of the olefin (co-)polymer composition finally obtained. The conditions for polymerization, particularly with respect to polymerization temperature and the quantity of the catalyst component (3) used, should be properly selected within the above-mentioned range.

The olefin (co-)polymer composition of the present invention comprises

mainly: 0.01 to 5 weight parts, preferably 0.02 to 2 weight parts, more preferably 0.05 to 1 weight parts of the olefin (co-)polymer (a) as the above-mentioned component (a); and 100 weight parts of the olefin (co-)polymer (b) as the component (b).

The amount of the olefin (co-)polymer (a) as the component (a) is preferably within the above-mentioned range in view of improvement in melt tension and crystallization temperature of the olefin (co-)polymer composition obtained, and also in view of homogeneity of the composition.

In the olefin (co-)polymer composition of the present invention, it is preferable that the melt tension (MS) measured at a temperature of 230 ℃ and the melt flow rate (MFR) measured under a load of 21.18 N at a temperature of 230 ℃ has a relationship as follows:

$$\log(\mathrm{MS}) > -0.76 \times \log(\mathrm{MFR}) + 0.45$$

Although there is no specific upper limit, if the melt tension is too high, the moldability of the composition is deteriorated. Therefore, it is preferably within a range of:

$$-0.76 \times \log(\mathrm{MFR}) + 2.00 > \log(\mathrm{MS}) > -0.76 \times \log(\mathrm{MFR}) + 0.45.$$

More preferably, it is within a range of:

$$-0.76 \times \log(\mathrm{MFR}) + 1.80 > \log(\mathrm{MS}) > -0.76 \times \log(\mathrm{MFR}) + 0.54.$$

The melt tension (MS) at 230 ℃ used herein is a value of tension (unit: cN) measured according to a process as follows: using MELT TENSION II (produced by TOYO SEIKI SEISAKU-SHO, Ltd.), an olefin (co-)polymer composition is heated to 230 ℃ in the apparatus, then the molten olefin (co-)polymer composition is extruded through a nozzle having a diameter of

2.095 mm into an atmosphere at a temperature of 23 ℃ at a rate of 20 mm/minute, thus producing a strand, and then the tension of the string-like olefin (co-)polymer composition, which is formed when the strand is taken up at a rate of 3.14 m/minute, is measured.

The olefin (co-)polymer composition of the present invention can be produced by any method as long as the melt tension of the composition is high enough to satisfy the above-mentioned range, but it is easier to produce it by a method comprising: preparing a preactivated catalyst by performing a preliminary activation of a polymerization catalyst comprising at least the catalyst components (1) and (2) by (co-)polymerizing olefins using the catalyst to form a small amount of an olefin (co-)polymer (a) having a particular intrinsic viscosity prior to main (co-)polymerization; and further combining the preactivated catalyst with the catalyst component (3) to produce a catalyst for main (co-)polymerization; and then (co)polymerizing olefins using the catalyst for main (co-)polymerization to form olefin (co-)polymer (b), thus obtaining the final product of the olefin (co-)polymer composition.

The preliminary activation treatment used in the method of producing the composition of the present invention is carried out under the following conditions:

The preliminary activation is carried out by forming 0.01 to 5,000 g, preferably 0.02 to 3,000 g of olefin (co-)polymer (a) per 1 g of the catalyst component (1) by supplying 0.01 to 10,000 g, preferably 0.02 to 8,000 g of olefins per 1 g of the catalyst component (1) either in the absence or the

presence of solvents of not exceeding 100 liters per 1 g of the catalyst component (1), in the presence of 0.001 to 5,000 millimole, preferably 0.01 to 1,000 millimole of a polymerization catalyst comprising the catalyst components (1) and (2) of the present invention with respect to Ti atoms contained in the catalyst component (1) per 1 liter of the olefin (co-)polymerization volume.

The term "polymerization volume" used in the present specification refers to the volume of the liquid phase in a polymerization reactor in the case of liquid phase polymerization, and to the volume of the gas phase in a polymerization reactor in the case of gas phase polymerization.

The amount of the catalyst component (1) used is preferably within the above-mentioned range, so that an efficient and controlled reaction rate of the olefin (co-)polymerization can be maintained. If the amount of the catalyst component (2) used is too small, the (co-)polymerization reaction rate is excessively slowed down. On the other hand, even if a larger amount of the catalyst component (2) is used, sufficient improvement of the (co-)polymerization reaction rate cannot be expected. Furthermore, if too large an amount of the solvent is used, a large reactor is required, and moreover, an efficient polymerization reaction rate is difficult to controll and maintain.

The preliminary activation can be carried out in the liquid phase using solvents. Examples of the solvents include aliphatic hydrocarbons such as butane, pentane, hexane, heptane, octane, isooctane, decane, dodecane and the like; alicyclic hydrocarbons such as cyclopentane, cyclohexane,

methylcyclohexane and the like; aromatic hydrocarbons such as toluene, xylene, ethylbenzene and the like; gasoline fraction or hydrogenized diesel oil fraction; and olefins themselves. Furthermore, the polymerization activation also can be carried out in the gas phase without using any solvents.

Although the preliminary activation may be performed in the presence of hydrogen, it is preferable not to use hydrogen in order to form a high molecular weight polyethylene (a) having an intrinsic viscosity [$\eta_a$] of 15 to 100 dl/g.

Other conditions for the preliminary activation are not particularly limited, as long as a certain amount of the high molecular weight olefin (co-)polymer (a) having an intrinsic viscosity [$\eta_a$] of 15 dl/g to 100 dl/g, preferably 17 dl/g to 50 dl/g can be produced. However, it is usually carried out at a relatively low temperature, about −40℃ to 50℃, preferably −40℃ to 30℃, more preferably −40℃ to 20℃, at a pressure of 0.1 MPa to 5 MPa, preferably 0.2 MPa to 5 MPa, more preferably 0.3 MPa to 5 MPa, for a period of from 1 minute to 24 hours, preferably from 5 minutes to 18 hours, more preferably from 10 minutes to 12 hours.

Furthermore, in a more preferable embodiment of the present invention, an additional preliminary activation is carried out before or before and after said preliminary activation to form 0.01 g to 100 g of an olefin (co-)polymer (aa) having an intrinsic viscosity [$\eta_{aa}$] lower than the intrinsic viscosity [$\eta_a$] of the olefin (co-)polymer (a) per 1 g of the catalyst component (1).

The additional preliminary activation is usually carried out at a temperature of –40℃ to 100℃, at a pressure of 0.1 MPa to 5 MPa, for a period of from 1 minute to 24 hours.

The types of the respective catalyst components, solvents and olefins, which are respectively used in the additional preliminary activation may be the same as those used in the preliminary activation.

In order to achieve the object of the present invention, it is more preferable that the olefin used as the main monomer in the additional preliminary activation is of the same type as the olefin used as the main monomer in the main (co-)polymerization.

The olefin (co-)polymer (aa) formed in the additional preliminary activation has an intrinsic viscosity [$\eta_{aa}$] within a range lower than the intrinsic viscosity [$\eta_a$] of the olefin (co-)polymer (a), and it is finally incorporated in the final product as a part of the olefin (co-)polymer (b) as the component (b) after the main (co-)polymerization.

When only the preliminary activation is carried out, whisker-like or lump type olefin (co-)polymer (a) may be formed depending on the conditions, causing problems in production such as adhesion of the polymer to the wall of the reactor for the preliminary activation, failure of ejection from the reactor for the preliminary activation, and also formation of a block type (co-)polymer during the main (co-)polymerization. Furthermore, a problem might occur that the olefin (co-)polymer (a) cannot be dispersed well in the olefin (co-)polymer (b) formed in the main (co-)polymerization, and as a result the melt tension of the olefin (co-)polymer composition obtained may

be not sufficiently improved. On the other hand, if the above-mentioned additional preliminary activation is also carried out, the shape of the catalyst slurry obtained can be improved, and therefore the problems in production are solved; and moreover, because the olefin (co-)polymer (aa) formed in the additional preliminary activation has an intrinsic viscosity $[\eta_{aa}]$ lower than the intrinsic viscosity $[\eta_a]$ of the olefin (co-)polymer (a) formed in the preliminary activation, the olefin (co-)polymer (aa) can be well-dispersed in the olefin (co-)polymer (b), so that the melt tension of the olefin (co-)polymer composition obtained as the final product can be improved.

Accordingly, it is more preferable that the intrinsic viscosity $[\eta_{aa}]$ of the olefin (co-)polymer (aa) is higher than the intrinsic viscosity $[\eta_b]$ of the olefin (co-)polymer (b) formed in the main (co-)polymerization.

The preliminarily activated catalyst or the catalyst treated further by the additional preliminary activation thus obtained is further combined with the above-mentioned catalyst component (3) so as to produce a catalyst for main olefin (co-)polymerization, and then it is provided for use in the main (co-)polymerization of olefins having 2 to 12 carbon atoms to produce the olefin (co-)polymer (b).

Furthermore, the catalyst component (2) may be additionally used at this time as needed.

The above-mentioned catalyst for main (co-)polymerization of olefin, including the catalyst component (2) added as needed in the main (co-)polymerization, comprises each component within the same range in quantity as the combination of the catalyst components in producing the

olefin (co-)polymer (b) as the component (b) of the present invention as specified above.

The catalyst component (2) additionally used in a catalyst for the main olefin (co-)polymerization as needed may be of either the same or different type as that used in the preliminary activation.

The catalyst for main (co-)polymerization of olefins can be used directly in the main olefin (co-)polymerization in combination with the catalyst component (3) and the catalyst component (2) added as needed after the above-mentioned preliminary activation. However, the catalyst for the main (co-)polymerization also may be prepared by combining the catalyst components (2) and (3) with a particulate product obtained by removing solvents, unreacted olefins, the catalyst component (2) and the like from the preactivated catalyst by filtration or decantation, or with a suspension produced by adding a solvent to the particulate product. Furthermore, the catalyst for the main (co-)polymerization also may be prepared by combining the catalyst components (2) and (3) with a particulate product obtained by removing solvents and unreacted olefins from the preactivated catalyst by vaporizing them by distillation under reduced pressure, inert gas flow or the like, or with a suspension produced by adding a solvent to the particulate product.

In the method of producing olefin (co-)polymer compositions of the present invention, the preactivated catalyst is used in an amount of 0.0001 to 5,000 millimole, preferably 0.001 to 1,000 millimole with respect to the transition metal atoms contained in the preactivated catalyst per 1 liter of

the polymerization volume. When the transition metal compound catalyst component is used in an amount within the above-mentioned range, an efficient and controlled reaction rate of the olefin (co-)polymerization can be maintained.

In the main (co-)polymerization of olefins using the above-mentioned preactivated catalyst in the present invention, the above-mentioned known polymerization processes and polymerization conditions, which are specified above in the production of the olefin (co-)polymer (b), are used.

In the method of producing an olefin (co-)polymer composition of the present invention, the polymerization conditions are chosen so that the amount of the olefin (co-)polymer (a) derived from the preactivated catalyst used becomes 0.01 to 5 weight parts, preferably 0.02 to 2 weight parts, more preferably 0.05 to 1 weight parts per 100 weight parts of the olefin (co-)polymer (b) formed in the main (co-)polymerization.

After the main (co-)polymerization is completed, after-treatment with known processes such as deactivating catalyst, removing catalyst residue, drying and the like is carried out as needed, so that an olefin (co-)polymer composition having an intended high melt tension and high crystallization temperature is finally obtained.

In the method of producing an olefin (co-)polymer composition of the present invention, an olefin (co-)polymer (a) having a high intrinsic viscosity is formed by preliminary activation, and then dispersed uniformly in the olefin (co-)polymer composition finally obtained. Therefore, it is possible to adjust the amount of the preactivated catalyst collectively as needed, and

moreover, because the main (co-)polymerization of olefins can be carried out by a normal olefin (co-)polymerization with a known process, the product can be obtained in the same amount as in usual olefin (co-)polymer production.

By adopting the method of producing an olefin (co-)polymer composition according to the present invention in which a preactivated catalyst is used, an olefin (co-)polymer composition satisfying the above-mentioned relationship between the melt tension (MS) at 230 ℃ and the melt flow rate (MFR) measured under a load of 21.18 N, at a temperature of 230 ℃ can be easily obtained.

The olefin (co-)polymer composition thus obtained is mixed with various types of additives such as antioxiants, ultraviolet ray absorbing agents, antistatic agents, nucleating agents, lubricants, flame retardants, anti-blocking agents, colorants, inorganic or organic fillers, or the like as needed, and further with various types of synthetic resins. Then, the mixture is usually heated and melt-mixed, and further cut into particulates to prepare pellets, which are then provided for producing various molded products.

Hereinafter the present invention will be explained in further detail with reference to examples and comparative examples.

Definitions of the terms and measurement methods used in the examples and the comparative examples are as follows:

(1) Intrinsic Viscosity [ $\eta$ ]: the value measured with an Ostwald's viscometer produced by Mitsui Toatsu Chemicals Inc. of the limiting viscosity measured in tetralin at a temperature of 135 ℃ (unit : dl/g).

(2) Melt Flow Rate (MFR): the value measured under the condition 14 in Table 1 according to JIS K7210 (under a load of 21.18 N, at a temperature of 230 ℃) (unit: g/10 minutes).

(3) Melt Tension (MS): the value measured with MELT TENSION II (produced by TOYO SEIKI SEISAKU-SHO, Ltd.) (unit: cN).

(4) Crystallization Temperature (Tc) : the value of the crystallization temperature was measured as follows: using a Differential Scanning Calorimetry VII (produced by PERKIN-ELMER, Ltd.), the temperature of an olefin (co-)polymer composition was raised from the room temperature to 230 ℃ at a rate of increase of 30 ℃/minute, the same temperature being maintained for 10 minutes. Then the temperature was lowered to −20 ℃ at a rate of decrease of −20 ℃/minute, the same temperature being maintained for 10 minutes. Then the temperature was raised to 230 ℃ at a rate of increase of 20 ℃/minute, the same temperature being maintained for 10 minutes. Then the temperature was lowered to 150 ℃ at a rate of decrease of −80 ℃/minute, and then the temperature was further lowered from 150℃ at a rate of decrease of −5 ℃/minute, while the temperature of the olefin (co-)polymer composition indicating the maximum peak of crystallization was determined as the value of the crystallization temperature (unit : ℃).

(5) Thermal stability: First, 0.1 weight parts of 2,6-di-t-butyl-p-cresol and 0.1 weight parts of calcium stearate were mixed with 100 weight parts of an olefin (co-)polymer composition. Then the mixture was melt-mixed and pelletized at 230 ℃ with an extruding pelletizer having a screw diameter of

40 mm to prepare pellets of the olefin (co-)polymer composition.

Then, using the initial pellets as obtained, final pellets were produced by repeating the melt-mixing and pelletizing with the above-mentioned extruding pelletizer twice. Then, with respect to the initial pellets and the final pellets, the melt flow rates (MFR) (unit: g/10 minutes) were respectively measured according to the condition 14 in Table 1 of JIS K7210, and the difference between the values of MFR of the pellets initially obtained and the final pellets (i.e. MFR of the final pellets − MFR of the initial pellets = △ MFR) was calculated.

It is shown that the smaller the value of the difference (△ MFR), the better the thermal stability.

(6) Rigidity: From the pellets of an olefin (co-)polymer composition prepared in (5), a test piece of JIS form was produced with an injection molding machine in conditions of the molten resin temperature at 230 ℃ and the die temperature at 50 ℃. Then, after being allowed to stand for 72 hours in a room having a humidity of 50 % and a temperature of 23 ℃, the bending elastic modulus of the test piece was measured in accordance to JIS K 7203 (unit: MPa).

Example 1

(1) Preparation of a Titanium Trichloride Composition (III):

Six liters of n-hexane, 5.0 moles of diethyl aluminum monochloride (DEAC), and 12 moles of diisoamyl ether were mixed at 25 ℃ for one minute, and then it was allowed to react at the same temperature for five minutes to obtain a reaction product ( I ) (diisoamyl ether/DEAC in a ratio of 2.4 by

mole). After 40 moles of titanium tetrachloride was put into a reactor in which the air was substituted with nitrogen gas, it was heated to 35 ℃, and then the total amount of the reaction product solution ( I ) was dropped therein over 180 minutes. The same temperature was maintained for 30 minutes, and then the temperature was raised to 75 ℃, and it was allowed to further react for one hour, and then it was cooled down to room temperature. Then, the supernatant was removed from the reaction mixture, and a process of adding 20 liters of n-hexane and removing the supernatant by decantation was repeated four times to obtain 1.9 kg of a solid product ( II ). 1.6 kg of diisoamyl ether and 3.5 kg of titanium tetrachloride were added to a suspention, prepared by dissolving the total amount of the solid product ( II ) in 30 liters of n-hexane at room temperature (20 ℃) in about one minute, and then it was allowed to react at a temperature of 65 ℃ for one hour. After the reaction was completed, it was cooled down to room temperature (20 ℃), and the supernatant was removed from the suspension by decantation. Then 40 liters of n-hexane was further added to the reaction mixture and stirred for 10 minute, and a process of removing the supernatant by allowing the suspension to stand was repeated five times, and then it was dried under reduced pressure to obtain a titanium trichloride composition (III). It was found out that 1 g of the titanium trichloride composition (III) contained 252 mg of Ti atoms.

(2) Preparation of a Preactivated Catalyst:

After substituting the air in a 100 liters capacity stainless steel reactor having an inclined-turbine agitator with nitrogen gas, 70 liters of n-hexane,

1.5 mole of diethyl aluminum monochloride, and 250 g (1.32 millimole with respect to Ti atoms) of the titanium trichloride composition (Ⅲ) prepared in (1) above were put into the reactor. Then, 800 g of propylene was supplied, and an additional preliminary activation was carried out at 5 ℃ for three minutes.

Separately, a polymer formed by a preliminary polymerization conducted under the same conditions above was analyzed, and as a result, it was found that 2 g of propylene per 1 g of the titanium trichloride composition (Ⅲ) was formed into a polypropylene (aa-1), and the polypropylene (aa-1) had an intrinsic viscosity [$\eta_{aa\text{-}1}$] of 3.0 dl/g measured in tetralin at 135 ℃.

After the reaction period was completed, unreacted propylene was discharged outside the reactor, and the gas phase in the reactor was replaced with nitrogen gas once. Then, while maintaining the temperature in the reactor at 5 ℃, ethylene was continuously supplied to the reactor for 4 hours so as to maintain the pressure inside the reactor at 0.59 MPa, thus carrying out the preliminary activation.

Separately, a polymer formed by a preliminarily activating polymerization conducted under the same conditions was analyzed, and as a result, it was found that 22 g of the polymer was present per 1 g of the titanium trichloride composition (Ⅲ), and the polymer had an intrinsic viscosity [$\eta_{T2}$] of 25.7 dl/g measured in tetralin at 135 ℃.

The amount ($W_2$) of the polyethylene (a-1) formed by the preliminary activation using ethylene per 1 g of the supported Ti containing catalyst

component is determined as the difference between the amount ($W_{T2}$) of the polymer formed through the preliminary activation per 1 g of the titanium trichloride composition (III) and the amount ($W_1$) of the polypropylene (aa−1) formed through the additional preliminary activation per 1 g of the titanium trichloride composition (III) calculated according to the following equation:

$$W_2 = W_{T2} - W_1$$

Furthermore, the intrinsic viscosity $[\eta_a]$ of the polyethylene (a) formed by the preliminarily activating polymerization using ethylene is determined from the intrinsic viscosity $[\eta_{aa\text{-}1}]$ of the polypropylene (aa−1) formed by the additional preliminary activation and the intrinsic viscosity $[\eta_{r2}]$ of the polymer obtained after the preliminary activation by calculating according to the following equation:

$$[\eta_a] = ([\eta_{T2}] \times W_{T2} - [\eta_{aa\text{-}1}] \times W_1) / (W_{T2} - W_1)$$

According to the above equations, it was found out that 20 g of the polyethylene (a) per 1 g of the titanium trichloride composition (III) was formed by the preliminary activation using ethylene, and the polyethylene (a) had an intrinsic viscosity $[\eta_a]$ of 28.0 dl/g.

After the reaction period was completed, unreacted ethylene was discharged outside the reactor, and the gas phase in the reactor was replaced with nitrogen gas twice. Then, 800 g of propylene was supplied, and an additional preliminary activation was carried out at 5 ℃ for three minutes.

Separately, a polymer obtained after an additional preliminary activation conducted under the same conditions was analyzed, and it was

found that 23.7 g of polymer was present per 1 g of the titanium trichloride composition (Ⅲ), and that the polymer had an intrinsic viscosity [$\eta_{T3}$] of 24.1 dl/g measured in tetralin at 135 ℃. Calculating in the same way as mentioned above, it was found out that the amount ($W_3$) of the polypropylene (aa-2) formed by the additional preliminary activation was 1.7 g per 1 g of the titanium trichloride composition (Ⅲ), and the polypropylene (aa-2) had an intrinsic viscosity [$\eta_{aa\text{-}2}$] of 3.4 dl/g.

After the reaction period was completed, unreacted propylene was discharged outside the reactor. Then, the gas phase in the reactor was replaced with nitrogen gas three times, thus producing a preactivated catalyst slurry for main (co-)polymerization.

(3) Production of a Polypropylene Composition (Main (co-)polymerization of Propylene):

After replacing the air in a 500 liters capacity stainless steel polymerization reactor having an agitator with nitrogen gas, 300 liters of n-hexane, 1.3 mole of diethyl aluminum monochloride, 0.09 mole of p-toluic acid methyl ester, and 17.1 g (0.09 mole with respect to Ti atoms) of the preactivated catalyst slurry obtained in (2) above as the titanium trichloride composition (Ⅲ) were put into the polymerization reactor at 20 ℃. Then, after introducing 500 liters of hydrogen into the reactor and raising the temperature to 70 ℃, propylene was continuously supplied into the reactor for 4 hours at a polymerization temperature of 70 ℃ so as to maintain the pressure of the gas phase in the reactor at 1.1 MPa, thus carrying out the main polymerization of propylene.

After the polymerization period was completed, 1 liter of methanol was introduced into the polymerization reactor, and catalyst deactivation was conducted at 70 ℃ for 15 minutes. Then, after discharging unreacted gas, removal of the solvents and drying of the polymer were conducted, so that 68.5 kg of polymer having an intrinsic viscosity [$\eta_T$] of 1.86 dl/g was obtained.

It was found out that the polymer obtained was a polypropylene composition containing 0.50 weight % of the polyethylene (a) formed by the preliminary activation as the component (a), and that the polypropylene component (b) had an intrinsic viscosity [$\eta_T$] of 1.73 dl/g.

0.1 weight parts of 2,6-di-t-butyl-p-cresol and 0.1 weight parts of calcium stearate were mixed with 100 weight parts of the polypropylene composition obtained. Then, the mixture was pelletized with an extruding pelletizer having a screw diameter of 40 mm at 230 ℃, thus producing pellets. According to evaluation by measuring the respective properties of the pellets obtained, it was found out that the pellets had a MFR of 5.0 g/10 minutes, a crystallization temperature of 125.8 ℃, and a melt tension (MS) of 3.1 cN.

Example 2

A polypropylene composition was produced using the same conditions as the Example 1 except that the conditions for the preliminary activation using ethylene were modified to change the amount of the polyethylene (a) formed, and an evaluation sample of Example 2 was prepared.

The properties of the polypropylene composition obtained are shown in

Table 1.

Comparative Example 1

A polypropylene composition was produced using the same conditions as in the Example 1 except that the preliminary activation of the titanium trichloride composition (Ⅲ) using ethylene was not carried out. The isotactic pentad tacticity of the polypropylene composition obtained was measured, and the result was 0.969. Furthermore, the molecular weight distribution of the polypropylene composition was measured, and the result was 7.3 for Mw/Mn. Based on these results, it is determined that the molecular weight distribution of the polypropylene (b) of the Example 1 is 7.3. Then, an evaluation sample of the Comparative Example 1 was prepared from the polypropylene composition obtained.

The evaluation results of the properties of the polypropylene composition obtained are shown in Table 1.

Comparative Example 2

A polypropylene composition was produced using the same conditions as the Comparative Example 1 except that p-toluic acid methyl ester was not used in the main polymerization of propylene and that the amount of hydrogen added was 200 liters. It was found out that the polypropylene composition obtained had an isotactic pentad tacticity of 0.947 and a molecular weight distribution (Mw/Mn) of 5.1. Furthermore, an evaluation sample of the Comparative Example 2 was prepared from the polypropylene composition obtained.

The evaluation results of the properties of the polypropylene

composition obtained are shown in Table 1.

Example 3

(1) Preparation of a Titanium Trichloride Composition (III):

After 27.0 moles of titanium tetrachloride were added to 12 liters of n-hexane and cooled down to a temperature of 1 ℃, 12.5 liters of n-hexane containing 27.0 moles of diethyl aluminum monochloride were dropped therein at 1 ℃ over four hours. After the dropping was completed, the mixture was allowed to react while maintaining the same temperature for 15 minutes. Then the temperature was increased to 65 ℃ over a period of one hour, and the reaction was further continued at the same temperature for one hour. Then, the supernatant was removed from the reaction mixture, and a process comprising 10 liters of n-hexane and removing the supernatant by decantation was repeated five times, and thus 5.7 kg of a solid product (II) was obtained. 1.8 kg of the solid product (II) obtained was suspended in 11 liters of n-hexane, and 1.8 liters of diisoamyl ether was added to the suspension. After the suspension was stirred at 35 ℃ for one hour, it was rinsed with 3 liters of n-hexane, which was repeated for a total of five times, so that a treated solid product was obtained. The treated solid product obtained was suspended in 6 liters of n-hexane solution containing 40 % by volume of titanium tetrachloride and 5 % by volume of silicon tetrachloride.

Then, the temperature of the suspension was raised to 65 ℃, and reaction was continued at the same temperature for two hours. After the reaction was completed, the solid product obtained was rinsed with 20 liters

of n-hexane, which was repeated for a total of three times, then dried under reduced pressure, so that a titanium trichloride composition (Ⅲ) was obtained. It was found out that 1 g of the titanium trichloride composition (Ⅲ) contained 265 mg of Ti atoms.

(2) Preparation of a Preactivated Catalyst:

After replacing the air in a 100 liters capacity stainless steel reactor having an inclined-turbine agitator with nitrogen gas, 70 liters of n-hexane, 1.5 mole of diethyl aluminum monochloride and 250 g (1.38 millimole with respect to Ti atoms) of the titanium trichloride composition (Ⅲ) prepared in (1) above were added into the reactor. Then, 800 g of propylene was supplied, and thus an additional preliminary activation was carried out at 0 ℃ for three minutes.

Separately, a polymer formed by a preliminary polymerization conducted under the same conditions as above was analyzed, and it was found that 1.7 g of propylene per 1 g of the titanium trichloride composition (Ⅲ) was formed into polypropylene (aa-1), and that the polypropylene (aa-1) had an intrinsic viscosity $[\eta_{aa\text{-}1}]$ of 3.4 dl/g measured in tetralin at 135 ℃.

After the reaction period was completed, unreacted propylene was discharged outside the reactor. Then, the gas phase in the reactor was replaced with nitrogen gas once, and while maintaining the temperature in the reactor at 0 ℃, ethylene was continuously supplied into the reactor for two hours so as to maintain the pressure in the reactor at 0.69 Mpa. Thus preliminary activation was carried out.

Separately, a polymer formed by a preliminary polymerization

conducted under the same conditions was analyzed, and it was found that 12 g of the polymer was present per 1 g of the titanium trichloride composition (III), and that the polymer had an intrinsic viscosity [$\eta_{T2}$] of 27.1 dl/g measured in tetralin at 135 ℃.

By calculating according to the same equations used in Example 1, it was found that 10.3 g of the polyethylene (a) per 1 g of the titanium trichloride composition (III) was formed by the preliminary activation using ethylene, and that the polyethylene (a) had an intrinsic viscosity [$\eta_a$] of 31.0 dl/g.

After the reaction period was completed, unreacted ethylene was discharged outside the reactor. Then, the gas phase in the reactor was replaced with nitrogen gas three times, thus preparing a preactivated catalyst slurry for main (co-)polymerization.

(3) Production of a Polypropylene Composition (Main (co-)polymerization of Propylene):

After replacing the air in a 500 liters capacity stainless steel polymerization reactor having an agitator with nitrogen gas, 300 liters of n-hexane, 1.2 mole of diethyl aluminum monochloride, 0.09 mole of 3-aminopropyltriethoxysilane, and 14.5 g (0.08 mole with respect to Ti atoms) of the preactivated catalyst slurry obtained in (2) above as the titanium trichloride composition (III) were added into the polymerization reactor at 20 ℃. Then, after introducing 550 liters of hydrogen into the reactor and raising the temperature to 72 ℃, ethylene was supplied into the reactor at a rate of 4 liters/minute, at a polymerization temperature of 72 ℃, and further

propylene was continuously supplied into the reactor for 4 hours so as to maintain the pressure of the gas phase at 1.1 MPa, thus carrying out main copolymerization of propylene and ethylene.

After the polymerization period was completed, 1 liter of methanol was introduced into the reactor, and catalyst deactivation was carried out at 70 ℃ for 15 minutes. Then, after discharging unreacted gas, the solvent was removed and the polymer was dried, so that 51.5 kg of a copolymer composition having an intrinsic viscosity [$\eta_T$] of 1.62 dl/g and containing 2.9 weight % of ethylene was obtained.

It was found out that the copolymer composition thus obtained was a propylene-ethylene copolymer composition containing 0.29 weight % of the polyethylene (a) formed by preliminary activation as the component (a), and that the propylene-ethylene copolymer as the component (b) had an intrinsic viscosity [$\eta_T$] of 1.53 dl/g.

0.1 weight parts of 2,6-di-t-butyl-p-cresol and 0.1 weight parts of calcium stearate were mixed with 100 weight parts of the copolymer composition obtained, and the mixture was pelletized with an extruding pelletizer having a screw diameter of 40 mm at 230 ℃, thus preparing pellets. The respective properties of the pellets were measured and evaluated, and it was found out that the pellets had a MFR of 11.5 g/10 minutes, a crystallization temperature of 109.2 ℃, and a melt tension (MS) of 1.7 cN.

Comparative Example 3

A polypropylene composition was produced using the same conditions

as the Example 3 except that preliminary activation of the titanium trichloride composition (Ⅲ) using ethylene was not carried out. The molecular weight distribution of the polypropylene composition obtained was measured, and the result was 6.8 for Mw/Mn. Accordingly, the molecular weight distribution of the propylene-ethylene (co-)polymer (b) in the Comparative Example 3 is determined as 6.8. Furthermore, an evaluation sample of the Comparative Example 3 was prepared from the (co-)polymer composition obtained. The respective properties of the copolymer composition obtained are shown in Table 1.

Table 1

| Nos. of the Examples and Comparative Examples | | Ex. 1 | Ex. 2 | Comp. Ex. 1 | Comp. Ex. 2 | Ex. 3 | Comp. Ex. 3 |
|---|---|---|---|---|---|---|---|
| (Note 1) PO (a) | Intrinsic Viscosity [η] (dl/g) | 28.0 | 28.0 | — | — | 31.0 | — |
| | Weight Parts | 0.50 | 0.15 | — | — | 0.29 | — |
| (Note 2) PO (b) | Intrinsic Viscosity [η] (dl/g) | 1.73 | 1.73 | 1.73 | 1.79 | 1.53 | 1.53 |
| | Molecular Weight Distribution (Mw/Mn) | 7.3 | 7.3 | 7.3 | 5.1 | 6.8 | 6.8 |
| | Weight Parts | 100 | 100 | 100 | 100 | 100 | 100 |
| Olefin (co-)polymer composition | Intrinsic Viscosity [η] (dl/g) | 1.86 | 1.77 | 1.73 | 1.93 | 1.62 | 1.53 |
| | Melt Tension (MS) (cN) | 3.1 | 1.7 | 0.5 | 1.2 | 1.7 | 0.3 |
| | Crystallization Temperature (Tc) (℃) | 125.8 | 124.0 | 122.5 | 118.6 | 109.2 | 106.0 |
| | MFR of the Initial Pellets (g/10 munutes) | 5.0 | 6.7 | 7.7 | 2.8 | 11.5 | 16.0 |
| | MFR of the Final Pellets (g/10 munutes) | 5.2 | 6.9 | 8.0 | 3.0 | 12.0 | 16.5 |
| | △MFR (g/10 minutes) | 0.2 | 0.3 | 0.3 | 0.2 | 0.5 | 0.5 |
| | Bending Elastic Modulus (MPa) | 1850 | 1780 | 1690 | 1420 | 1390 | 1270 |

Note 1: PO (a) represents the olefin (co-)polymer (a).
Note 2: PO (b) represents the olefin (co-)polymer (b).

Industrial Applicability

As shown in the Examples above, because the olefin (co-)polymer composition of the present invention has a high melt tension, high crystallization temperature and high rigidity, it is excellent in moldability. Furthermore, because it has excellent thermal stability, it exhibits high productivity in molding processes, and moreover, it can be melted again after being used as a molded product, and then recycled for producing another molded product. It is particularly suitable for blow molding, foaming and extrusion molding, however, it is not limited to these molding fields, and can be provided for producing various molded products, for example, various containers such as blow molded bottles and the like, films, sheets, pipes, fibers and the like, by injection molding, T-die molding, heat molding or the like. Thus, the fields in which olefin (co-)polymers can be used is dramatically expanded.

# CLAIMS

1. An olefin (co-)polymer composition mainly comprising:

   (a) 0.01 to 5 weight parts of an olefin (co-)polymer (a) having an intrinsic viscosity [$\eta_a$] of 15 to 100 dl/g measured in tetralin at 135℃; and

   (b) 100 weight parts of an olefin (co-)polymer (b) having an intrinsic viscosity [$\eta_b$] of 0.2 to 10 dl/g measured in tetralin at 135℃, which is produced by (co-)polymerizing olefins using a polymerization catalyst comprising catalyst components (1), (2) and (3) as follows:

   Catalyst Component (1): a titanium trichloride composition (Ⅲ)

   Catalyst Component (2): an organic aluminum compound ($A_2$)

   Catalyst Component (3): at least one compound selected from the group consisting of aromatic carboxylic acid esters (E), and organic silicon compounds (S) containing at least one organic group selected from the group consisting of a Si-O-C group and a mercapto group.

2. A composition according to claim 1, wherein the titanium trichloride composition (Ⅲ) is produced by reacting titanium tetrachloride with either an organic aluminum compound ($A_1$) or a reaction product ( I ) of the organic aluminum compound ($A_1$) and an electron donor ($B_1$) to form a solid product (Ⅱ), and then reacting the product (Ⅱ) with an electron donor ($B_2$) and an electron acceptor.

3. A composition according to claim 1, wherein the olefin (co-)polymer composition has a relationship between melt tension (MS) at 230 ℃ and melt flow rate (MFR) measured under a load of 21.18 N at 230 ℃ as follows:

$$\log (MS) > -0.76 \times \log (MFR) + 0.45.$$

4. The composition of claim 3, wherein the relationship between MS and MFR is as follows:

$$-0.76 \times \log (MFR) + 2.00 > \log (MS) > -0.76 \times \log (MFR) + 0.45.$$

5. The composition of claim 3, wherein the relationship between MS and MFR is as follows:

$$-0.76 \times \log (MFR) + 1.80 > \log (MS) > -0.76 \times \log (MFR) + 0.54.$$

6. A composition according to claims 1, wherein the olefin (co-)polymer (a) is either an ethylene homopolymer or an ethylene-olefin (co-)polymer containing not less than 50 weight % of ethylene polymerization units.

7. A composition according to claim 1, wherein the olefin (co-)polymer (b) is either a propylene homopolymer or a propylene-olefin copolymer containing not less than 50 weight % of propylene polymerization units.

8. A composition according to claim 1, wherein a ratio (Mw/Mn) of the weight average molecular weight (Mw) to the numerical average molecular weight (Mn) of the olefin (co-)polymer composition is in the range of 6 to 30.

9. A method of producing an olefin (co-)polymer composition comprising: performing a preliminary activation of a polymerization catalyst comprising catalyst components (1) and (2) below by (co-)polymerizing olefins to form 0.01 to 5,000 g of an olefin (co-)polymer (a) having an intrinsic viscosity [$\eta_a$] of 15 to 100 dl/g measured in tetralin at 135℃ per 1 g of the catalyst component (1) to prepare a preactivated catalyst; combining the preactivated catalyst further with a catalyst component (3) below to prepare a polymerization catalyst; and then performing (co-)polymerization of olefins using the polymerization catalyst to form an olefin (co-)polymer (b) having an intrinsic viscosity [$\eta_b$] of 0.2 to 10 dl/g measured in tetralin at 135℃, thus producing an olefin (co-)polymer composition mainly comprising:

(a) 0.01 to 5 weight parts of an olefin (co-)polymer (a) having an intrinsic viscosity [$\eta_a$] of 15 to 100 dl/g measured in tetralin at 135℃; and

(b) 100 weight parts of an olefin (co-)polymer (b) having an intrinsic viscosity [$\eta_b$] of 0.2 to 10 dl/g measured in tetralin at 135℃:

Catalyst Component (1): a titanium trichloride composition (III)

Catalyst Component (2): an organic aluminum compound ($A_2$)

Catalyst Component (3): at least one compound selected from the group consisting of aromatic carboxylic acid esters (E), and organic silicon compounds (S) containing at least one organic group selected from the group consisting of a Si-O-C group and a mercapto group.

10. A production method according to claim 9, wherein the titanium trichloride composition (Ⅲ) is produced by reacting titanium tetrachloride with either an organic aluminum compound ($A_1$) or a reaction product ( I ) of the organic aluminum compound ($A_1$) and an electron donor ($B_1$) to form a solid product (Ⅱ), and then reacting the product (Ⅱ) with an electron donor ($B_2$) and an electron acceptor.

11. A production method according to claim 9, wherein the (co-)polymerization of olefins is performed using a preactivated catalyst further combined with an additional catalyst component (2) below:

Catalyst Component (2): an organic aluminum compound ($A_2$).

12. A production method according to claim 9, wherein the olefin (co-)polymer composition obtained has a relationship between melt tension (MS) at 230 ℃ and melt flow rate (MFR) measured under a load of 21.18 N at 230 ℃ as follows:

$$\log (MS) > -0.76 \times \log (MFR) + 0.45.$$

13. A production method according to claim 12, wherein the relationship between MS and MFR is as follows:

$$-0.76 \times \log (MFR) + 2.00 > \log (MS) > -0.76 \times \log (MFR) + 0.45.$$

14. A production method according to claim 12, wherein the relationship between MS and MFR is as follows:

$$-0.76 \times \log(MFR) + 1.80 > \log(MS) > -0.76 \times \log(MFR) + 0.54.$$

15. A production method according to claim 9, wherein the olefin (co-)polymer (a) is either an ethylene homopolymer or an ethylene-olefin (co-)polymer containing not less than 50 weight % of ethylene polymerization units.

16. A production method according to claim 9, wherein the olefin (co-)polymer (b) is either a propylene homopolymer or a propylene-olefin copolymer containing not less than 50 weight % of propylene polymerization units.

17. A production method according to claim 9, wherein an additional preliminary activation is performed by (co-)polymerizing olefins before or both before and after the preliminary activation to form 0.01 to 100 g of an olefin (co-)polymer (aa) having an intrinsic viscosity of $[\eta_{aa}]$ lower than the intrinsic viscosity $[\eta_a]$ of the olefin (co-)polymer (a) formed by the preliminary activation per 1 g of the catalyst component (1).

18. A production method according to claim 9, wherein the olefin (co-)polymer (aa) is either a propylene homopolymer or a propylene-olefin copolymer containing not less than 50 weight % of propylene polymerization units.

19. A production method according to claim 9, wherein a ratio (Mw/Mn) of the weight average molecular weight (Mw) to the numerical average molecular weight (Mn) of the olefin (co-)polymer composition is in the range of 6 to 30.

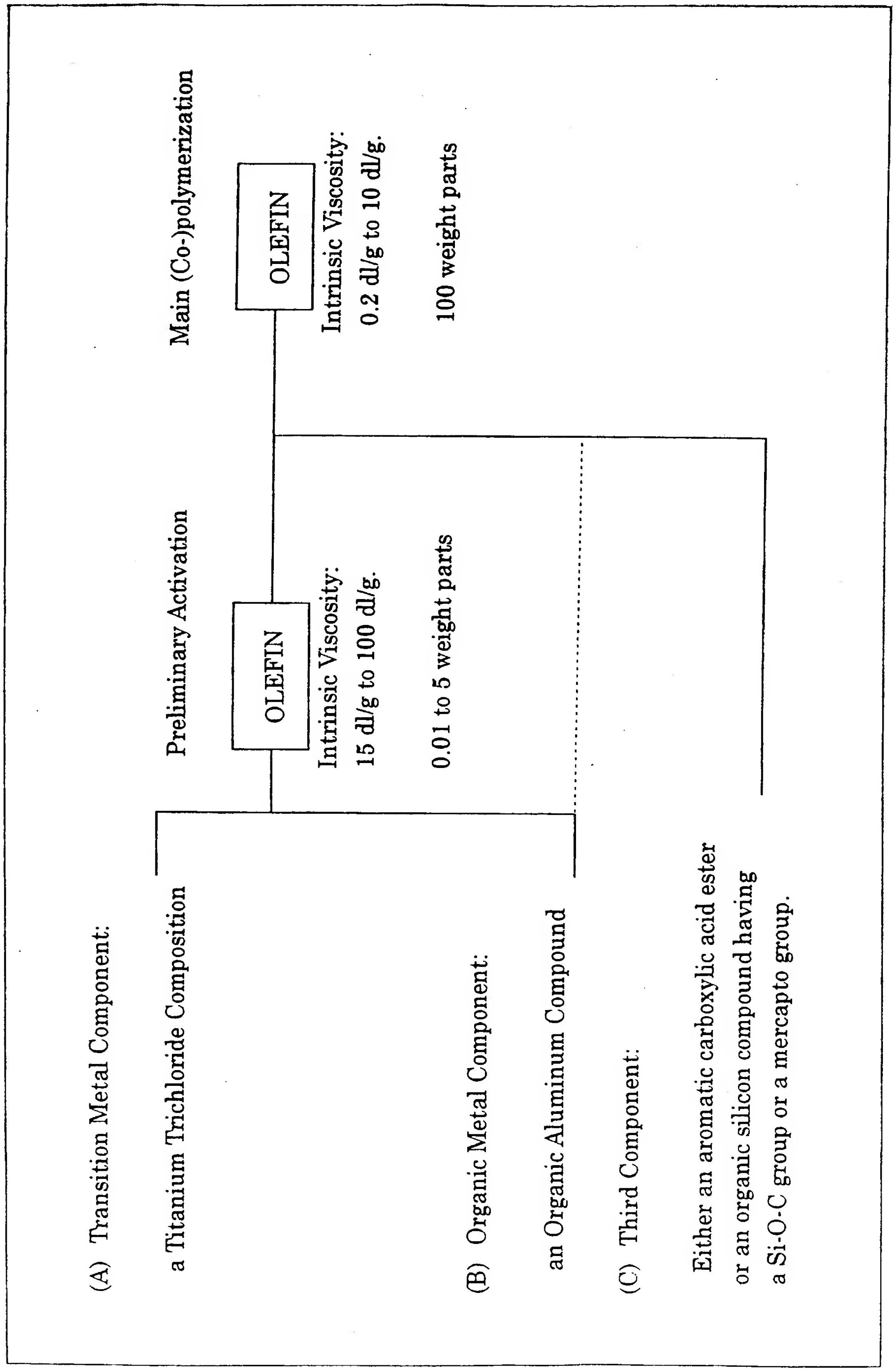
Main (Co-)polymerization
OLEFIN
Intrinsic Viscosity:
0.2 dl/g to 10 dl/g.
100 weight parts
Preliminary Activation
OLEFIN
Intrinsic Viscosity:
15 dl/g to 100 dl/g.
0.01 to 5 weight parts
(A) Transition Metal Component:
a Titanium Trichloride Composition
(B) Organic Metal Component:
an Organic Aluminum Compound
(C) Third Component:
Either an aromatic carboxylic acid ester
or an organic silicon compound having
a Si-O-C group or a mercapto group.

*FIG. 1.*

# INTERNATIONAL SEARCH REPORT

Inte onal Application No
PCT/JP 98/01603

**A. CLASSIFICATION OF SUBJECT MATTER**
IPC 6 C08L23/10 C08F10/00

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 C08L C08F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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| Date of the actual completion of the international search | Date of mailing of the international search report |
|---|---|
| 1 July 1998 | 09/07/1998 |

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|---|---|
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# INTERNATIONAL SEARCH REPORT

Inte onal Application No
PCT/JP 98/01603

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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